Magnetic Circular Dichroism in the X-ray Absorption Spectra of the CMR Compound, Yb₁₄MnSb₁₁

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This work is part of ongoing investigations into the magnetic and electronic properties of the rare-earth transition metal Zintl phases $A_{14}MnPn_{11}$ (A = Eu, Yb; Pn = Sb, Bi) at the Advanced Light Source. We have recently obtained exciting new results from X-ray magnetic circular dichroism (XMCD) investigations of the Yb₁₄MnSb₁₁ system. Specifically, we have used XMCD as an element specific probe into the nature of the magnetic moment in this system with the intention of exploring the proposed half-metallic nature of this compound and its related substitutional analogues. Our XMCD measurements indicate that Yb₁₄MnSb₁₁ is a half-metallic ferromagnet, and we have submitted our results for publication to Physical Review Letters.

The term half-metallic ferromagnet arises from theoretical predictions made by R.A. de Groot et al based on band structure calculations of the ferromagnetic Heusler alloy NiMnSb. 1 These calculations proposed a new phase of matter that displays separate electronic properties for majority-spin and minority-spin electrons. Specifically, one electron spin population is metallic and the other is insulating. Such a material, (possessing 100% spin polarization of the conduction electron) would hold significant technological potential as a single-spin electron source for spintronic devices, data storage applications, and high efficiency magnetic sensors. 2

The materials we are studying are new compounds that belong to a class of materials called transition-metal Zintl phases. These compounds are isostructural to $Ca_{14}AlSb_{11}$ and crystallize in the tetragonal space group $I4_1$ / acd (Z=8). The $Yb_{14}MnSb_{11}$, $Yb_{14}MnBi_{11}$ and $Eu_{14}MnSb_{11}$ analogues are each reported to order ferromagnetically at 56 K, 58 K and 28 K, and 92 K, respectively. $^{3-6}$ $Eu_{14}MnBi_{11}$ is an antiferromagnet with a Néel transition at $T_N=32$ K. 6 Each of these materials exhibits a large resistance drop associated with their unique magnetic ordering temperature. This behavior is attributed to colossal magnetoresistance effects, and could help support the proposal made by Pickett and Singh of a correlation between half-metallicity and colossal magnetoresistance. These systems are ideal for investigations into the links between magneto-resistance, magnetic moment and half-metallic behavior.

The ability to perform X-ray magnetic circular dichroism experiments on the EPU has allowed us to probe the dichroic characteristics of Mn and Sb in the Yb₁₄MnSb₁₁

system during experiments recently performed on beamline 4.0 of the ALS. Figure 1 shows the results from XMCD experiments on the Mn L₂₃, Sb M₄₅, and Yb N₄₅ edges of Yb₁₄MnSb₁₁. A dramatic dichroism effect is shown in the Mn L₂₃ region which is confined to one sub-component of the Mn edge and closely matches theoretical models for Mn²⁺, d⁵ dichroism (Figure 1d). The difference in intensity upon change of helicity is greater than 30%, and is strong evidence of a significant moment being present on the Mn. In contrast, no dichroism was observed in the Yb edges, but a small antialigned moment is present in the Sb M_{45} edges as shown on the left side of Figure 1. This result is surprising because initial models predicted that dichroism would be restricted to the Mn L₂₃ region (with no dichroism in the Sb M₄₅ region) and that it would be Mn³⁺, d^4 like in character. However, an ongoing collaboration with theoretical groups in the physics departments of the University of California, Davis and the University of Illinois, Urbana to model the Ca and Ba analogues of this structure type has now produced calculated results consistent with our experimental observations of how the Sb behaves in this system. They argue that the Mn should be in a 2+, d^5 configuration, and the Sb₄ cage surrounding the Mn should have a hole antialigned to the Mn moment giving a total moment of $\sim 4 \mu_B$ / formula unit. Our experimental results are consistent with these new theoretical results, and in addition, these comparisons of data with theoretical calculations and SQUID magnetometry measurements confirm that Yb14MnSb11 is indeed a halfmetallic ferromagnet.8-10

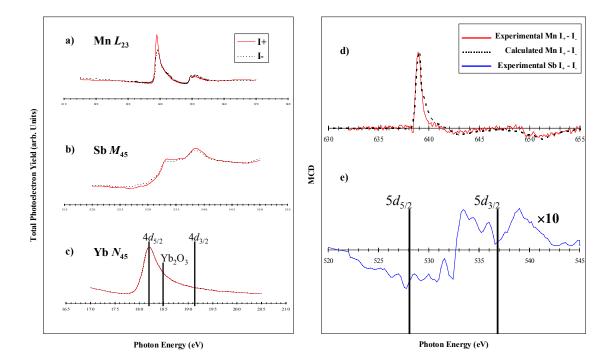


Fig. 1. The raw absorption spectra at plus and minus light polarization for a) Mn L_{23} , b) Sb M_{45} , and c) Yb N_{45} are shown on the left. The MCD spectra for d) the experimental Mn L_{23} denoted by a solid red line and the calculated Mn^{2+} , d^5 L_{23} denoted by a dashed black line, and e) the experimental Sb M_{45} denoted by a solid blue line. ¹⁰

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